

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Appl. No. : 09/625,710 Confirmation No. 6545
Applicants : Alfred E. Keller
Filed : July 25, 2000
For : Process for Producing Syngas in a
Short Contact Time Reactor Using
Catalytic Partial Oxidation of Hydrogen Sulfide

TC/A.U. : 1764
Examiner : Alexa A. Doroshenk

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DECLARATION OF ALFRED E. KELLER

Commissioner for Patents
P.O. Box 1450
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Sir:

PURPOSE OF DECLARATION

1. This Declaration is to establish the deficiency of at least the following reference as prior art against the above-identified U.S. patent application:

U.S. Patent No. 5,720,901 (*DeJong et al.*)

U.S. Patent No. 5,512,260 (*Kiliany et al.*)

U.S. Patent No. 5,676,921 (*Heisel et al.*)

STATEMENT OF FACTS

2. I, Alfred E. Keller, have a Bachelor of Science Degree in Chemical Engineering (1981) and a Bachelor of Science Degree in Chemistry (1981) from the Colorado School of Mines, and have practiced in the field of Gas Treating and Sulfur Recovery technology for over 19 years. I am currently employed as Consultant, Treating and Sulfur Processing, Downstream Technology for ConocoPhillips, Ponca City, Oklahoma. In the course of such employment, in addition to other duties, I am responsible for catalyst and reactor development and process design in the area of SynGas technology and sulfur recovery technology as well as process design and trouble shooting for amine systems, sour water handling systems, caustic treating, light olefins alkylation, Claus sulfur recovery units, and Claus plant tail gas treating units. I am the inventor of the above-identified patent application. I have extensive knowledge and experience in the areas of the design, synthesis and testing of catalysts and equipment

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for use in the petroleum and natural gas refining industry. I am familiar with the level of education and skill that was possessed by one of ordinary skill in the art working in the field of synthesis gas production from sulfur-containing hydrocarbon streams at the time of the above-identified invention.

3. I have read the specification and claims in the above-identified pending patent application and am familiar with the invention described therein. I have also read the above-listed patents and understand their teachings.

4. I am also familiar with catalytic partial oxidation processes for converting H_2S -containing light hydrocarbon feeds to synthesis gas. I am familiar with Claus and modified Claus sulfur recovery processes and with low-temperature (*i.e.*, below about $500^\circ C$) direct oxidation processes for converting H_2S to elemental sulfur and water. I am familiar with the behavior of hydrogen sulfide-containing light hydrocarbon gas streams, and with the behavior of elemental sulfur in its gas, liquid and solid phases.

5. Attached to this Declaration is an excerpt from a standard engineering reference book (the "GPSA handbook") that is widely used by chemical engineers working in the field of industrial sulfur recovery operations. This reference is more particularly identified as: Section 22, Sulfur Recovery; and Fig. 22-20 Vapor Pressure of Sulfur (Gas Processors Suppliers Association, ENGINEERING DATA BOOK, FPS Version, Vol. II, Sections 16-26, Eleventh Ed., published by Gas Processors Association, (1998), Tulsa, Oklahoma, p. 22-1 and 22-19).

6. Fig. 22-20 in the GPSA handbook shows the vapor pressure of sulfur versus temperature. The curve shows the vapor pressure of sulfur vapor that is in equilibrium with saturated sulfur liquid. The vapor pressure of sulfur is dependent only on the temperature of the liquid. In order for sulfur vapor to condense from the vapor phase to the liquid phase, the sulfur in the vapor phase must be in equilibrium with sulfur in the liquid phase. According to Raoult's Law, this occurs where the mole fraction of the sulfur vapor times the total system pressure is equal to the vapor pressure of the sulfur liquid times its mole fraction or:

$$Y_S P_T = X_S P_S^{sat} \quad (1)$$

In the immediate invention and the cited references, the combination of low P_T of the system and the gas components of the system involved make the gas phase act nearly ideally hence the use of the Raoult's law simplification.

7. Simplifying further, at the conditions nominally expected in a process according to equation (1), the left side of the equation is merely the partial pressure of the sulfur vapor. Sulfur is the only component of the mixture that would be expected to condense out of the mixture at the temperature of operation of a conventional sulfur condenser, ca. 260-375°F. Under those conditions, the mole fraction of sulfur in the liquid would be very close to 1. The right side of the equation is then the vapor pressure for sulfur vapor above a saturated sulfur liquid. Thus liquid sulfur could only be present when the partial pressure of sulfur in the vapor phase is equivalent to or greater than the vapor pressure of a saturated sulfur liquid.

8. Applying equation 1, above, at the conditions of *DeJong et al.*, the total sulfur content of the incoming feed hydrocarbon stream is 100 ppm or less. The nominal process of *DeJong et al.* is:



Using pure oxygen and methane contaminated with the specified amount of sulfur, the concentration of the elemental sulfur in the product, if any, would be at most (1.5/3) or 1/2 of the original feed composition as there are twice as many moles of gas in the products as in the feed gas. The maximum amount of the sulfur in the product would be $0.5(100) \times 10^{-6}$ or 50×10^{-6} moles sulfur/total moles gas. In mole fraction, this would be 0.00005. Operating at a total pressure of 1 atm., the partial pressure by Raoult's law would be 0.00005×1 or 0.00005 atm. This is assuming that the sulfur is present as S_1 . From the upper curve, and under sulfur condenser operation conditions, the partial pressure of sulfur needed to condense any sulfur liquid is between .001 and .01 atm. Since the partial pressure expected in the product stream in *DeJong et al.* would be expected to be less than 0.00005 atm, and since this is below the vapor pressure of saturated sulfur liquid, then the equilibrium relationship between liquid and vapor could not be established and no liquid would form. One of ordinary skill in the art would not even consider trying to condense the sulfur as a liquid from this stream.

9. A person of ordinary skill in the art at the time of the invention would be aware that sulfur vapor and liquid at nominal sulfur plant condenser conditions will be mostly S_8 with some S_6 . The partial pressures of the S_8 and S_6 vapor would be much smaller than for S_1 . Also, the vapor pressure would be smaller by about the same factor. This relationship is represented by the lower curve. So even though the lower curve shows condensation would take place at a lower vapor pressure, the partial pressure of the sulfur vapor would change accordingly. Again, one of ordinary skill in the art would recognize that sulfur could not be condensed from the product of *DeJong et al.*

10. Although operating at any higher pressure than 1 atm would increase the sulfur vapor partial pressure at the stated concentration in *DeJong et al.*, the total pressure of the system would need to increase to about 12-20 atm (176-294 psia) to get the partial pressure of sulfur vapor equal to the vapor pressure of saturated sulfur at the lowest practical sulfur condensation temperatures, ca. 265-275 F., where the greatest amount of sulfur would be removed from the product. One of ordinary skill in the art generally would design a sulfur condenser utilizing boiling water as a cooling medium. A steam pressure is normally chosen to be greater than process side pressure to prevent the contamination of the steam system with hydrocarbons or sulfur bearing compounds should the condensing tube leak. Water boiling at 177 psia boils at about 372 F, about 100 F higher than the desired condensation temperature for the greatest sulfur removal from the product from *DeJong et al.* Again, one of ordinary skill in the art would recognize the sulfur in the product from *DeJong et al.* could not be practically condensed in a typical sulfur condenser.

11. A substantially sulfur-free product, such as the substantially desulfurized synthesis gas exiting *DeJong et al.*'s desulfurization unit 22 by way of line 24, could not result from a sulfur condenser because of the inherent limitations of sulfur condensers, as discussed above. Therefore, one of ordinary skill in the art would know that a sulfur condenser could not be substituted for desulfurization unit 22 in the apparatus of *DeJong et al.*

12. If one of ordinary skill in the art were given the *DeJong et al.* and *Kiliary et al.* references, it would not occur to that person to even try to combine the teachings of those references at least because of the marked differences in the feeds, products and reactors. In addition, the knowledge that extremely low levels of sulfur as taught by *DeJong et al.* would not even be condensable would dissuade the ordinarily skilled worker from attempting such combination. There could be no reasonable expectation of obtaining useful sulfur as a result of such combination.

13. Similarly, the means for recovering elemental sulfur described by *Heisel et al.* is also incompatible with the apparatus of *DeJong et al.* for essentially the same reasons as discussed above with respect to *Kiliary et al.* The different oxidation reactors, different feeds, and different products would lead one of ordinary skill in the art away from trying to combine the apparatus. There would have been no reasonable expectation at the time of the above-identified invention of having a condensable amount of elemental sulfur in the product of the *DeJong et al.* process; therefore, the use

of a sulfur condenser instead of a conventional syngas desulfurization unit would have been considered pointless.

TIME OF PRESENTATION OF THE DECLARATION

14. This Declaration is submitted with the Response to the Office Action dated June 30, 2003, and is for the purpose of overcoming one or more ground of rejection or requirement made in that Office Action.

DECLARATION

15. I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Respectfully submitted,

Full name: ALFRED E. KELLER
Signature: *Alfred E. Keller*
Date: 8/21/03

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